PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Application No.:	09/918,158
Filing Date:	July 30, 2001
Applicant:	Robert A. Dichiara Jr.
Group Art Unit:	1731
Examiner:	Christopher A. Fiorilla
Title:	OXIDE BASED CERAMIC MATRIX COMPOSITES
Attorney Docket:	7784-000146

Director of the United States Patent and Trademark Office
P.O. Box 1450
Alexandria, VA 22313-1450

DECLARATION UNDER 37 C.F.R. § 1.131

Sir:

I hereby declare under penalty of perjury as follows:

1. That I am the sole inventor of the above-identified application.

2. That the invention was conceived and at least partially reduced to practice in this country prior to February 24, 1994, the filing date of the United States Patent No. 5,422,331 to Galligan et al. and prior to December 20, 1996, the filing date of the United States Patent No. 5,958,583 to Rorabaugh et al. In addition, this invention was

conceived and at least partially reduced to practice in this country prior to December 15, 1999, the filing date of United States Patent No. 6,497,776 to Butler et al., and prior to March 3, 1998, the filing date of United States Patent No. 6,110,439 to Deshpande et al.

3. I am the author of the notebook whose cover page is attached at Exhibit A. Pages from this notebook are attached as Exhibits B and C and the information contained within this notebook was either prepared by myself or under my direction.

4. That the invention was conceived and/or reduced to practice prior to February 25, 1994, as evidenced by the notebook page attached as Exhibit B. Exhibit B illustrates at least the initial conception and reduction to practice of a composition embodied by at least claim 1. A second page from the notebook is attached as Exhibit C and shows reduction to practice of a further embodiment of the invention claimed in at least claim 1 prior to February 25, 1994.

5. That the invention has never been abandoned, suppressed, or concealed.

6. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements are being made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false

statements may jeopardize the validity of the application, and patent issuing thereon, or any patent to which this verified statement is directed.

Dated: 8/25/04

Robert A. Dichiara, Jr.

(5)

Department [illegible]

Subject CML (M-106)

Name R. D. Oliva

Address

National Brand 43-649

Laboratory
Research Notebook

Dennison National Company, Holyoke, MA 01041

11" x 9¼" 4 x 4 Quad.
200 Sheets - 100 Sets

0 73333 43649 5

...ok M-106

Exhibit A

Robert DiChiara

DATE [redacted]

Mul-I-2

Charge #: IR-53704

VARIABLES		
A	Mullite (Baikalox Submicron Al_2O_3/SiO_2: 1.75)	Silica Sol/Mul 1.3

LAMINATE SIZE: 6.5" (Warp direction) x 9.5" (Fill direction) untrimmed
 # Plies: 8

a) Heat clean a piece of 8HS Nicalon fabric 39" wide (Fill direction) by 13" long (Warp direction).
b) Make up mix and use just after mixing or remix before use. Put 500 grams of Silica Sol (Nalco 2327) into the small ball mill and add 375 grams of submicron Baikalox Mullite (Al_2O_3/SiO_2: 1.75) mix for 0.5 hours. Add 1.5 grams of Dow Corning antifoam 1410. Mix for 3.5 hours use mixture after this point. If observe foaming when opening up mill or viscosity not right for prepreging let Bob D. know. If cannot use that day, ball mill mixture again for 2 hours before use.
c) Hand prepreg the fabric try and achieve about 33 % fiber to matrix ratio and lay-up 8 wet plies nested together, 6.5" (Warp direction) x 9.5".
d) **Fabric Wt.** 127.7 **g,**
Prepreg Wt. (actual) 307.1 g.
Fiber/Matrix ratio = Fabric Wt./Prepreg Wt. (actual) x 100 = 41.6 %.
e) Press cure the laminate
Apply 200 psi immedately and heat press to 200°F hold for 1/2 hour.
Heat press to ~~220~~°F at 1°F/min and hold for 1/2 hour.
Heat press to ~~350~~°F at 1°F/min. and hold for 1/2 hour.
Remove laminate ~~for post curing.~~ 2000°F
f) Post cure: To ~~1500°F~~/2 hr at 5 to 10°F/ minute.
g) Do physicals: % Porosity, % Matrix and % Fiber.
h) Cut laminate into flexure samples (0.5" x 5.5"[Warp direction]).
i) Heat treat samples placing samples into furnace at-temperature and remove to room temperature.

See below

m)

Test Samples:	Heat Treated	Testing Temperature
4 Flexure	None	RT
~~4 Flexure~~	~~1500°F/1hr~~	~~1500°F~~
4 Flexure	2000°F/1hr	2000°F
~~3~~ 4 Flexure	~~2000~~ 2400°F/1hr	RT

May want to Repeat with HfO_2

There is a key here for other work as well. Good demo laminate. #1 Submicron material #2 viscosity slurry #3 Bleeder package

May want to try submicron mullite in phosphate chemistry as well. See if submicron is major effect on viscosity

COMMENTS:
Program - hold was paused Set at 185 until 7:50pm went to 210°F for 1/2 hr. Then to 425°F / 1°F/min hold 1 hour. On 10-7-93 7:30AM removed from 245° press. Panel looked excellent, uniform surface and .105" thick seems hard. Post cured to 2000°F for 2 hrs. Part hardens up within 5 minutes at ~180°F Mixture putties-up after one day in a closed container

IRAD93

10/4/93

Exhibit B

Robert DiChiara

DATE: 2-5-97

Alum/Sol-I-2

Charge #: IR-569030

VARIABLES			
A	Alumina (SM-8)	B. Silica Sol/Alumina	1.3
C.	Nextel 610 (alumina fiber)		

LAMINATE SIZE: **6.5" (Warp direction) x 9.5" (Fill direction)** untrimmed
 # Plies: 8

a) Use heat cleaned Nextel 610 (8HS fabric) 39" wide (Fill direction) by 13" long (Warp direction).
b) Make up mixture and use just after mixing or remix just before use. Put 500 grams of Silica Sol (Nal 2327) into the small ball mill and add 375 grams of submicron Baikalox SM-8 (Al_2O_3) mix for 0.5 hou Add 1.5 grams of Dow Corning antifoam 1410. Mix for 3.5 hours use mixture after this point. If observe foaming when opening up mill add 0.5 grams of antifoam 1410, tumble for 5 minutes and recor on sheet. If problems or viscosity not right for prepreging let Bob D. know. If cannot use that day, ball mill mixture again for 2 hours before use.

If slurry still appears good from Alum/Sol-I-1 you can use slurry again if you high shear at 5000 rpm's for 5 minutes (please make note on sheet of the material you used).
c) Hand prepreg the fabric try and achieve about **38-40 % fiber to matrix ratio** and lay-up 8 wet pli nested together , 6.5" (Warp direction) x 9.5".
d) **Fabric Wt.** 108.8 **g,**
Prepreg Wt. (actual) 244.7 g.
Fiber/Matrix ratio = Fabric Wt./Prepreg Wt. (actual) x 100 = 44.2 %.
e) Make up cork dam set up with bleaderlease C on both sides like used in Mul-IV-1.
f) Press cure the laminate ,
use one layer of armolon and one layer of pink release glass on both sides
Apply 200 psi immedately and heat press at 2°F.min. to 180°F hold for 1.5 hour.
Heat press to 210°F at 1°F/min and hold for 1 hour.
Heat press to 425°F at 5°F/min. and hold for 1 hour.
Remove laminate for post curing.
g) Post cure: To 2000°F/2 hr at 5 to 10°F/ minute.
h) Reinfiltarate with SiO_2 Sol. (Nalco 2327) 2 times.
i) Weigh panel to start and reinfiltate panel with Silica Sol under vacuum (30" Hg) for 30 min.. Remove panel from sol and place in oven at 300°F for 30 min.. Pull panel out of oven and when at room temperature wipe off excessive silica powder off of surface and weigh. Repeat process 2 times. Weight as made 136.5g, 1st infiltration 141.2 g, 2nd infiltration 143.7g,
j)Fire panel to 2000°F for 2 hours. After firing _____g.
k)Cut panel in half and dry at 220°F.
l) Reinfiltarate half the panel with SiO_2 Sol. (Nalco 2327) for 2 more times.
m) Weigh panel to start and reinfiltate panel with Silica Sol..
Weight as made _____g, 3rd infiltration _____g, 4th infiltration _____g,
n) Fire panel to 2000°F for 2 hours. After firing _____g.
o) Do physicals: % Porosity, % Matrix and % Fiber.
p) Cut laminate into flexure samples (0.5" x 5.5"[Warp direction]).
q) Heat treat samples placing samples into furnace at-temperature and remove to room temperature.

r) Mechanical test both 2 and 4 times infiltarted panels.

Test Samples:	Heat Treated	Testing Temperature
4 Flexure	None	RT
4 Flexure	1800°F/1hr	1800°F

Exhibit C

COMMENTS: yields 800 gms of slurry after ball milling or 480 ml